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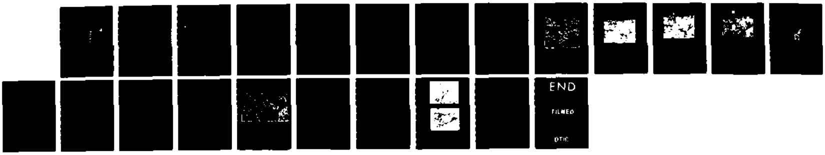

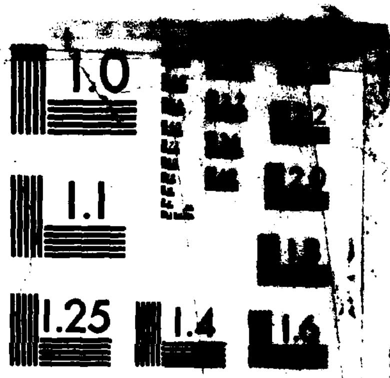

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Because this project relies on the joint participation of Research Triangle Institute (RTI) and University of North Carolina personnel, it is closely tied to the academic year. Delays in finding a graduate student suitable for the project prompted a request for a no-cost extension of the first year's work, which was granted. The completion of the first part of the project is on schedule with the extended period of performance. The work performed to date will be described in this report.

STUDY OF MEAN FREE PATH EFFECTS
ON GROWTH OF ULTRAFINE METALLIC AEROSOLS

AFOSR Contract Number F49620-84-C-0017
RTI Project Number 473U-2867

<u>Statement of Progress</u>

INTRODUCTION

This is a three-year project for investigating the dynamics of aerosol formation in gaseous atmospheres ranging from conditions at the earth's surface to those in the extreme upper atmosphere. The research involves theoretical and experimental determination of the behavior of ultrafine aerosol particles at high concentrations. The normal diffusion-limited coagulation and growth of aerosols is expected to be strongly modified as the particle diameter and the interparticle separation approach the mean free path of the gas molecules.

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THEORETICAL APPROACH TO THE PROBLEM

Very early in the program, a computer coagulation model was adapted for use at reduced pressures. The (unpublished) model had been developed in conjunction with an EPA effort on the growth of submicron aerosols from combustion sources. The model represents an adaptation of the coagulation mechanisms from the molecular regime (long mean free path) to the continuum regime (short mean free path), with the whole range covered in a consistent fashion.

The model was adapted for use on IBM-PC and other microcomputers and was used for generating limits on the experimental conditions that would be encountered. The details of the model are being written for publication at this time, and the microcomputer programs will be made available at the time of publication. It is somewhat surprising to note that the model runs almost as fast on the IBM-PC with coprocessor as on the UNIVAC mainframe computer on which is was developed. Certainly, with the interactive nature of the program for the personal computer, the overall throughput of the program can be much higher.

The model is based upon assumptions that are applicable during the early part of the experimental growth processes, namely spherical particles, which may coalesce as liquids or form spherical clusters of particles. As the later stages of growth occur, the resulting particles can easily depart from the spherical model, and another approach becomes necessary.

The Brownian coagulation kernel of Fuchs is used in the calculations, with corrections made for the molecular slip. Because of this approach, it is to be expected that the ultimate size distribution for continuum regime particles (mean free path much less than particle diameter) should approach the results of Friedlander. Figure 1 shows the results for the continuum regime, based upon two different versions of the model.

In the simpler but more time-consuming model, equal particle volumes are used to perform the calculations of the transitions from one particle size to another. In the faster model, the particle volumes are determined by a constant ratio of diameters, resulting in a model which requires volume corrections for each transition.

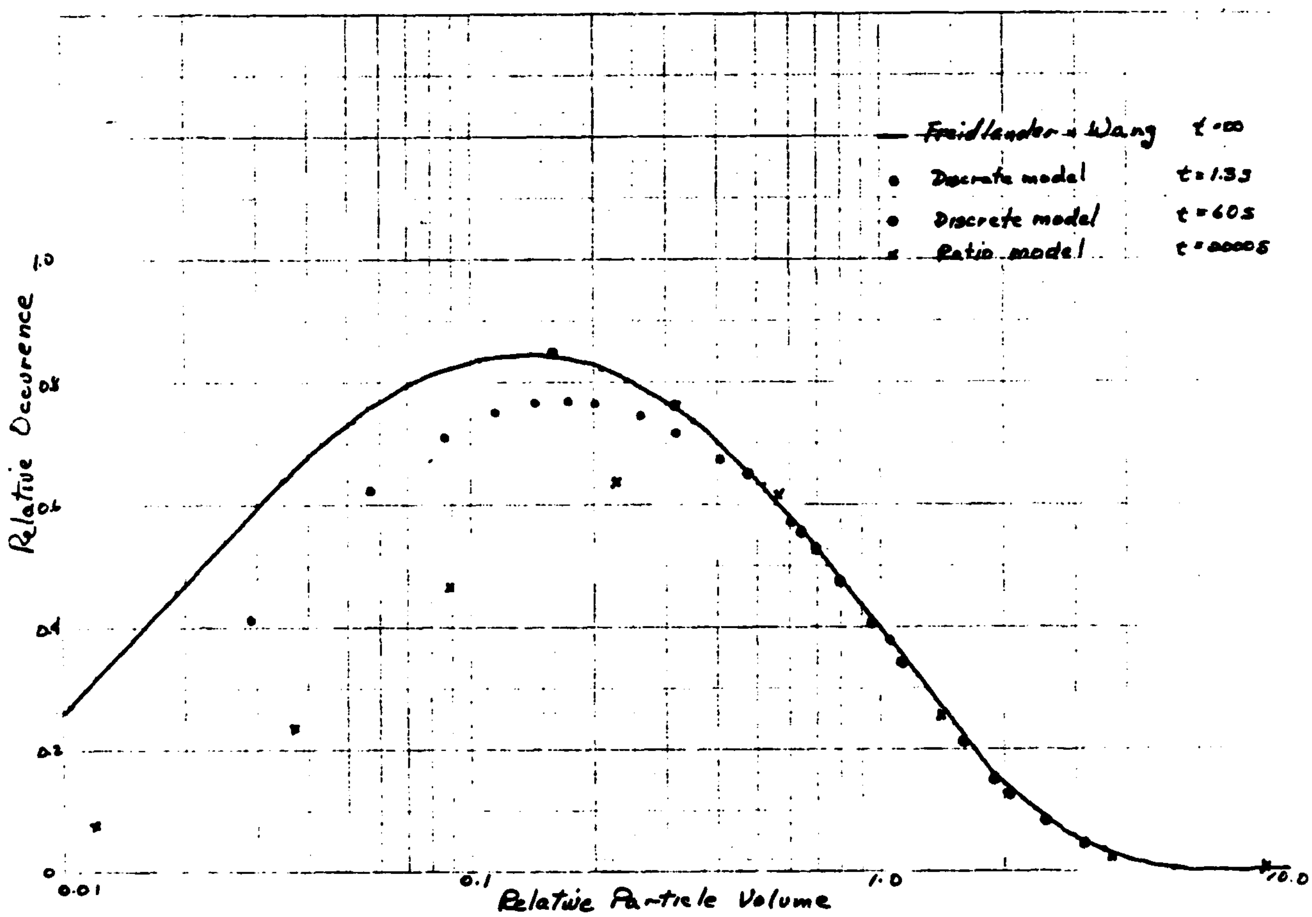

Figure 1. Comparison of discrete volume model and diameter ratio model with Friedlander's results for the continuum regime.

As can be seen, the equal volume model matches the results of Friedlander quite well, giving confidence that it is performing the calculations correctly. The ratio of diameters model does not do as well until the bin diameters produced closely spaced volume increments. Nonetheless, it allows a rapid evaluation of the parameters of a particular growth process and can serve as a screening tool for the equal volume model.

Even though the Brownian kernel strictly applies only to liquids, we are considering means of incorporating a void fraction into the resulting particles. This would allow the model to continue to be useful for the growth of solid particles, so long as the roughly spherical shape was maintained. There is some utility in doing this, provided the programs do not become unwieldy, since we think that the stages of growth with particle diameters less than the gas mean free path should continue in a roughly spherical manner.

For the description of the later stages of growth, we are devoting time and effort to a fractal description of the growth. Fractals are mathematical entities which have been developed to describe the physical irregularities of sytems. It is a new branch of mathematics, finding applications in numerous situations.

Our approach is to use a fractal description with theoretical growth models to match the fractal descriptions of the experimentally grown particles. There are several reasons for this approach. First is that the experimental results show the particles deviating strongly from a spherical shape, a condition that makes a purely theoretical treatment very difficult. Second, the fractal description of particle shapes appears to be a robust process, one insensitive to sampling of particle populations. Since our observations will necessarily be on small numbers of particles, a robust analysis will be very useful. Third, the theoretical models can be formulated in fairly general terms, so that as new interactions between particles become important, they can be included without undue mathematical complexity.

Another reason for addressing the fractal approach is that it has potential utility in describing the optical scattering properties of aerosols. Although we are not directly addressing this problem, it is to be expected that fractal descriptions of the scattering process will become available and, thus, allow our measurements to be tied in to other efforts.

For a brief description of the fractal analysis, we consider an arbitrary planar shape, such as in Figure 2 outlined by the heavy line. The figure is then masked with a plane-covering pattern (regular triangles, squares, pentagons, or hexagons) of different scales until the figure is either completely covered or one pattern covers the figure exactly. The number of cells that is required to cover the figure then depends on the dimension of each cell: the smaller the cell, the more are required. If the figure exhibits certain fractal characteristics, then a logarithmic plot of the number required to cover versus the length of the covering cell will give a straight line, whose slope can be used to express the fractal dimension. The region of straightness will be between the limits described above.

That is, the slope of the line will be:

$$k = d\ln(N)/d\ln(b), \tag{1}$$

where N is the number of covering squares, b is the length of the side of the square, and k will be termed the fractal dimension.

Figures 2 through 5 show the process for three sizes of squares, illustrating one of the intuitive advantages of a fractal description: the basic shape of the particle is preserved at all scales of coverage.

In the theoretical sense, a purely fractal object will have no lower limit of scale, and multiple objects can be combined to produce fractal objects of larger scales.

The field of fractal mathematics is so new that the techniques for using it are not well established. For instance, the more common description of fractal dimension of a planar object relies on determining the perimeter of the object as a function of step size. We intend to use the covering square method because it will integrate more easily with the image analysis equipment available for this project, but we did verify that the two methods give nearly equivalent results.

Using a random number generator, artificial particles were drawn by a computer using a spherical unit size. One such particle is shown in Figure 6. The fractal dimension of this object was determined by the two methods--perimeter and covering square. The fractal dimension resulting from the

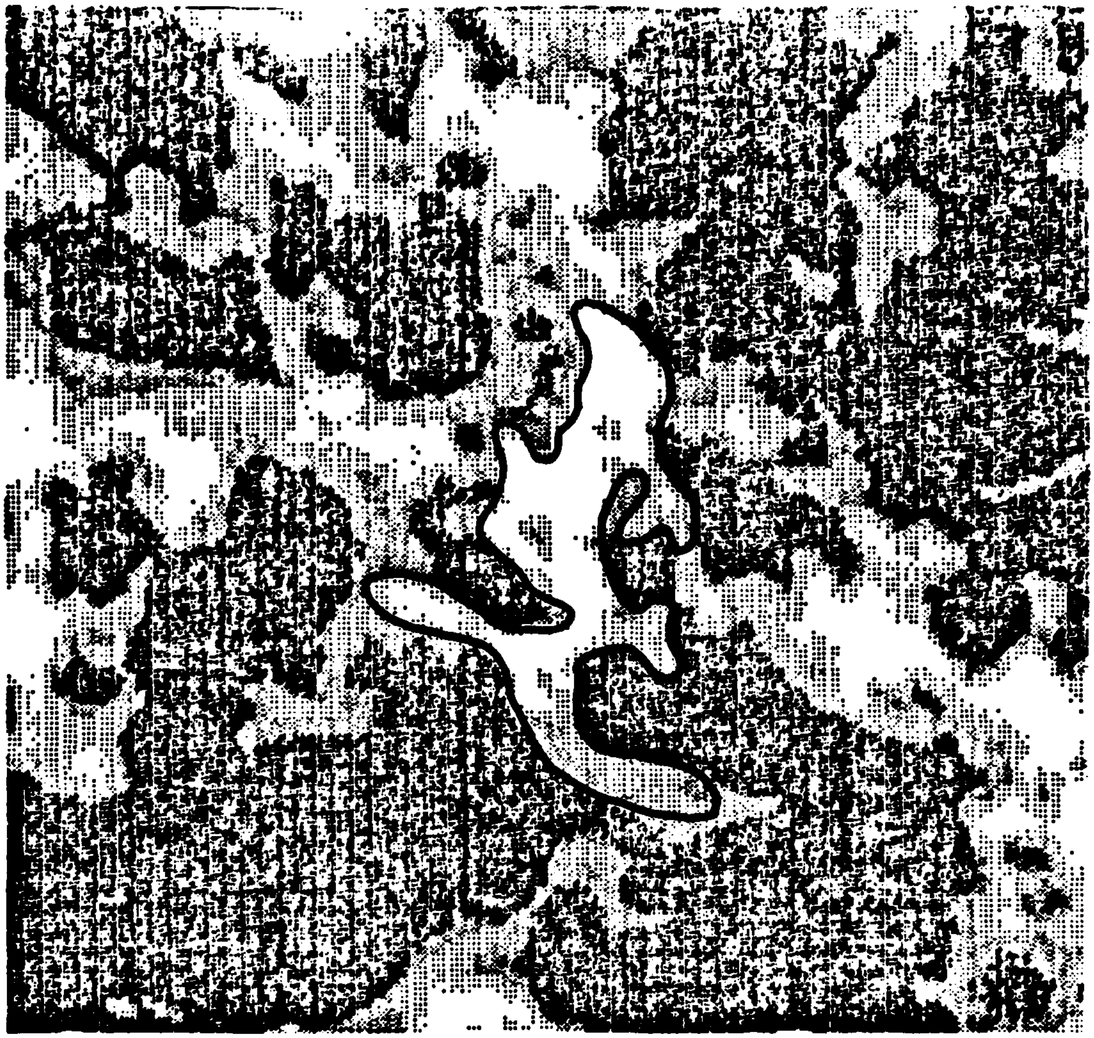

Figure 2. Shape representing an image of a particle for fractal analysis.

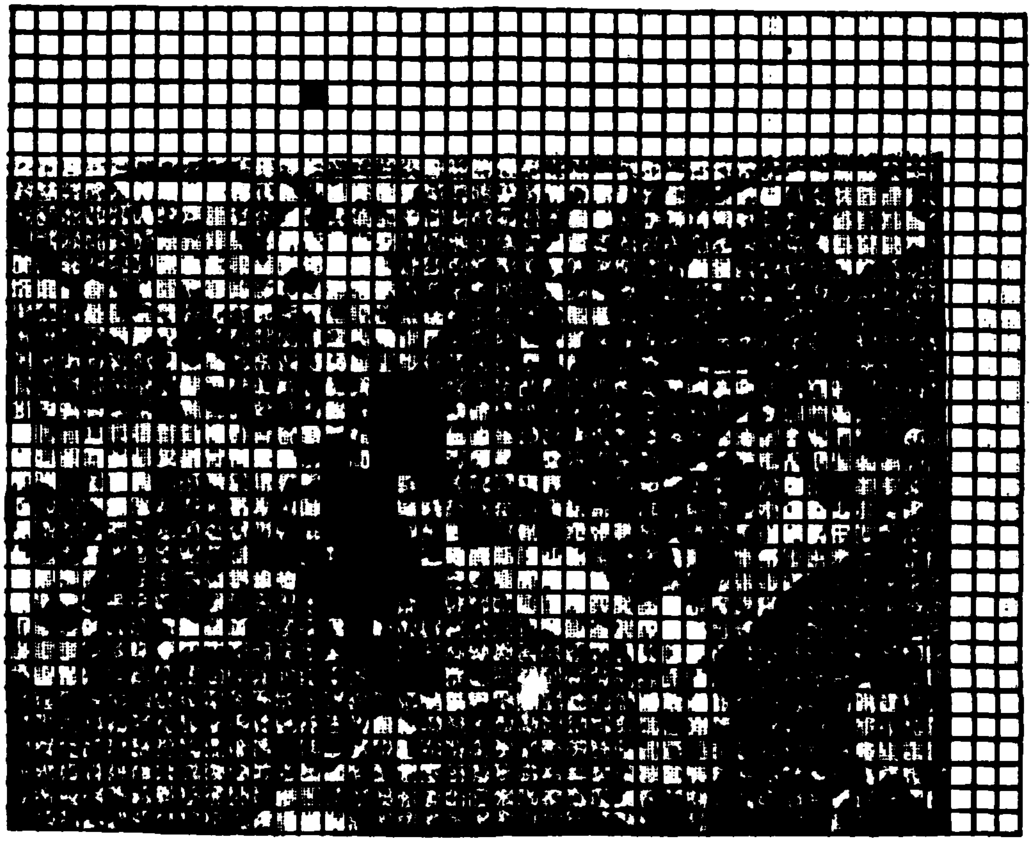

Figure 3. Tiling of shape by unit squares.

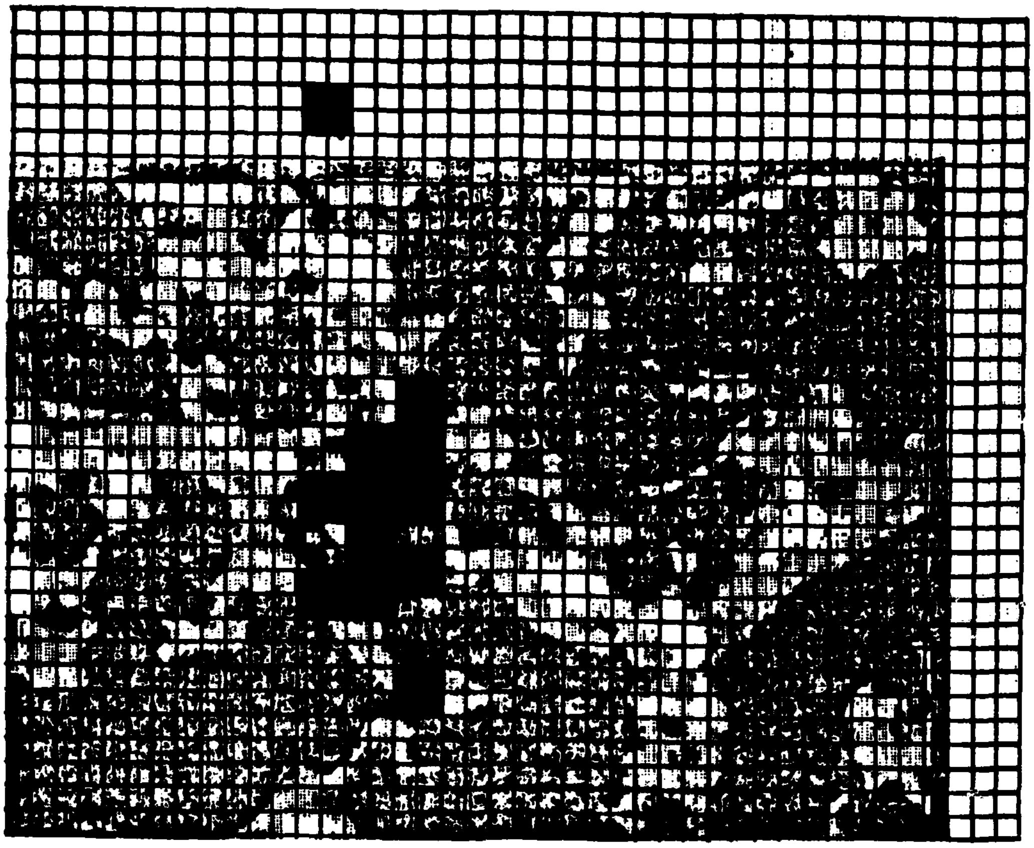

Figure 4. Tiling of shape by squares of Side 2.

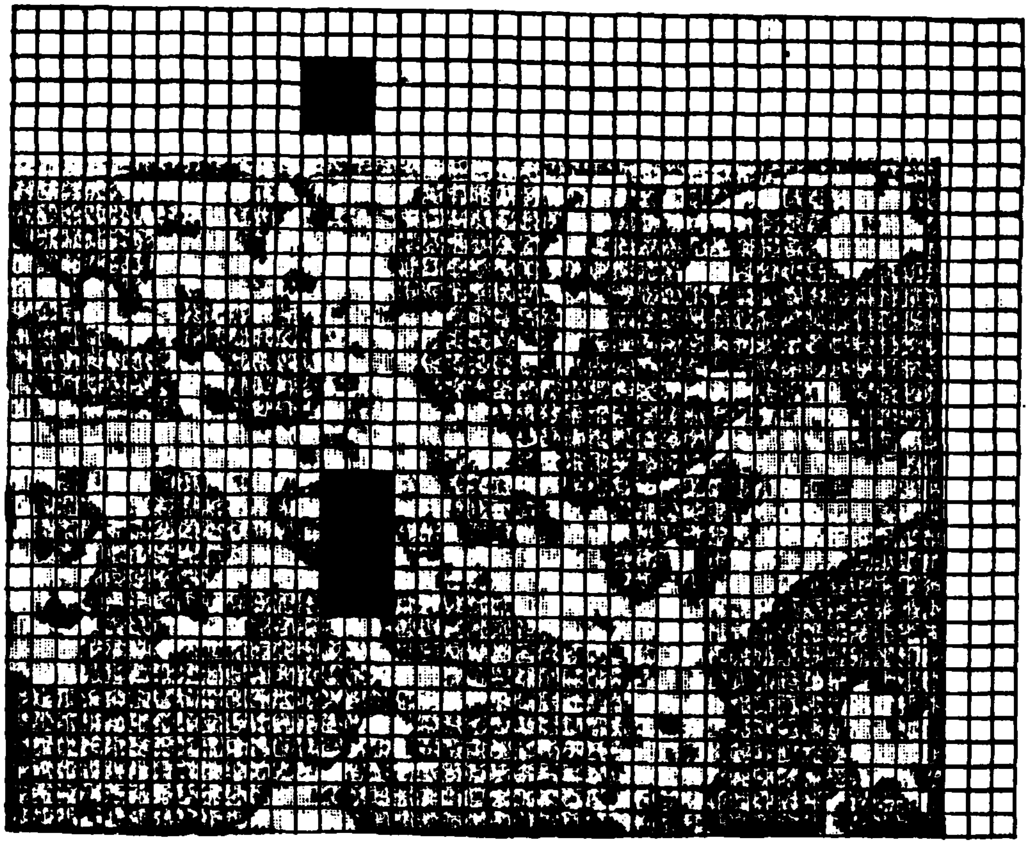

Figure 5. Tiling of shape by squares of Side 3.

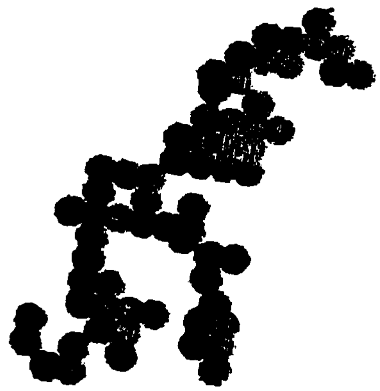

Figure 6. Computer generated particle made of agglomerated spheres.

perimeter technique was 1.53 and that resulting from the square technique was 1.68. The differences are considered minor at this stage, since the selection critieria for each method involve subjective judgments. Moreover, other particles generated in similar fashion showed as much particle-to-particle variation.

EXPERIMENTAL APPARATUS

A major part of the effort has been devoted to contructing a low pressure chamber for the experiments. The exploding wire generator used for aerosol production serves as a base for the chamber. The rest of the chamber was built around a large stainless-steel pipe tee. The pumping and pressure measuring system are interfaced to the tee through vacuum-tight flanges on the arms of the tee. This proved to be a flexible, yet rugged way to have access to the active volume of the chamber.

Since the minimum pressures expected from the experiments are within the range of a mechanical vacuum pump, modest vacuum techniques (gum rubber gaskets, for instance) sufficed for most of the connections. The system pressure is being measured by a capacitance manometer, which is adequate for all but the lowest pressures. When those pressures require measurement, a McLeod gauge will be used for absolute calibrations of the capacitance gauge.

An argon laser has been placed in position for light-scattering measurements with the chamber, but as yet the detection optics have not been mounted. They are being adapted from the optics of a white-light particle counting instrument; although they are optically very good, making them vacuum-tight has been a problem. Although the light scattering from irregular particles in this geometry is not well-defined, we intend to use the measurements to monitor the progress of the coagulation in a noninvasive manner.

An extractive sampling cylinder has been placed within the large chamber for capturing particles at various times during the growth phase. Consideration of the difficutlties of measuring particles (or even capturing particles) without disturbing the gas pressure led us to the extractive technique. The particles can be quickly raised to atmospheric pressures and flushed through several sampling devices.

We are presently using a laser particle counter for a qualtitative description of the particle size distribution, filter samples for the electron microscope observations, and a particle mass monitor for the conservation of mass within the chamber. Plans are under consideration for static sampling of the settling particles during the course of the growth.

Some of the particle size distributions measured by the counter are shown in Figures 7 and 8, for aerosol coagulating at two different pressures. Each figure shows the shape of the distribution at increasing times after the explosion, with the characteristic shift to larger particle sizes and broadening of the size spectrum.

For analyzing the electron micrographs and the theoretically produced particles, we have obtained a video camera and image analyser board for the PC. The software for obtaining the fractal dimensions of the particles is currently under development. The analysis package will form a routine part of the data reduction when it is fully in place.

The digitized picture can be printed as a variable density image on a dot-matrix printer, as seen in Figure 9. Although the printed image contains no more information than either the original photograph or the digital image itself, it can be reproduced fairly accurately on standard copying machines.

The digital information contains pixel information at a density of about 4000 per square inch. Since the image of the more interesting particles can be optically expanded before digitizing to cover an area of roughly 20 square inches on the screen, the basic fractal cells can span a linear range of about 300 to 1. The particles themselves, based upon our first observations, probably will not have that wide a range of characteristic dimensions, and so the analysis will not be limited by the measuring devices.

EXPERIMENTAL RESULTS TO DATE

The earliest experiments were devoted to obtaining a consistently metallic aerosol. It quickly became clear that the explosion of the wires would cause oxidation of the particles unless steps were taken to prevent that occurrence. The use of a pure nitrogen gas atmosphere has proved adequate to this time. Initially, it was also thought desirable to use a noble metal for

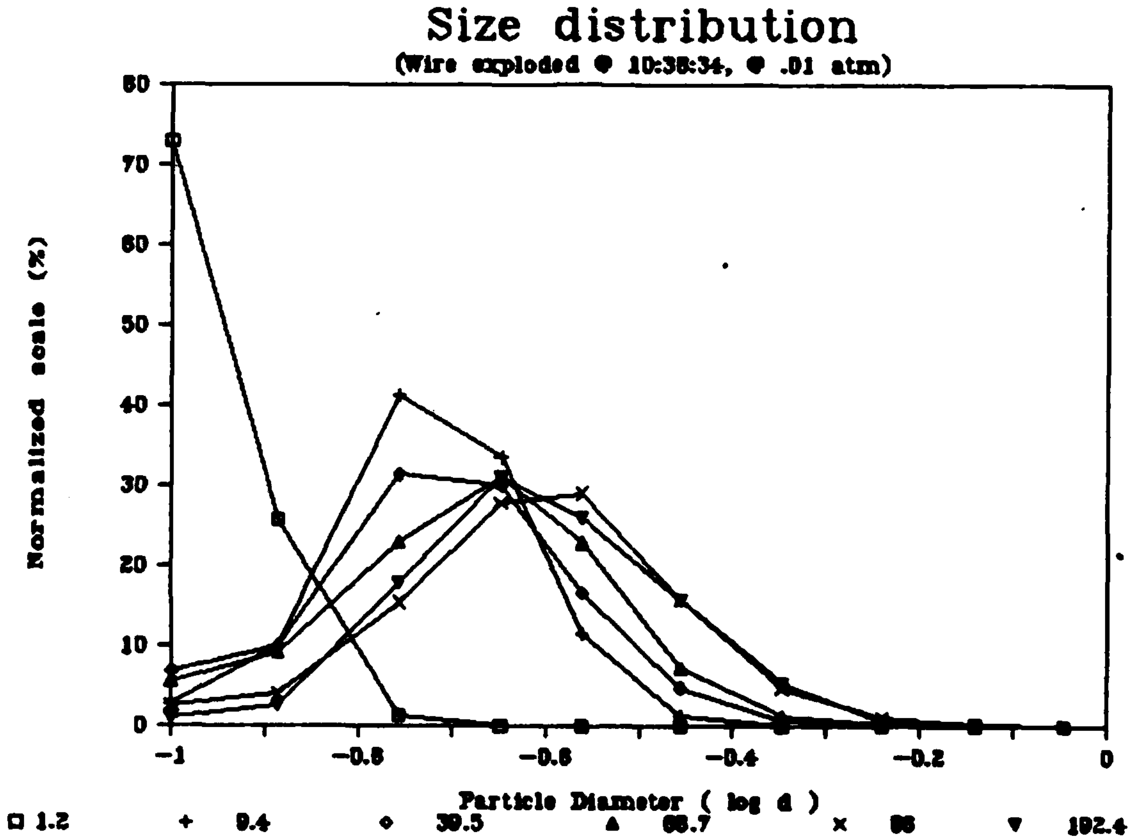

Figure 7. (Upper) Silver aerosol coagulated at 0.01 atmosphere for different times (values in seconds).

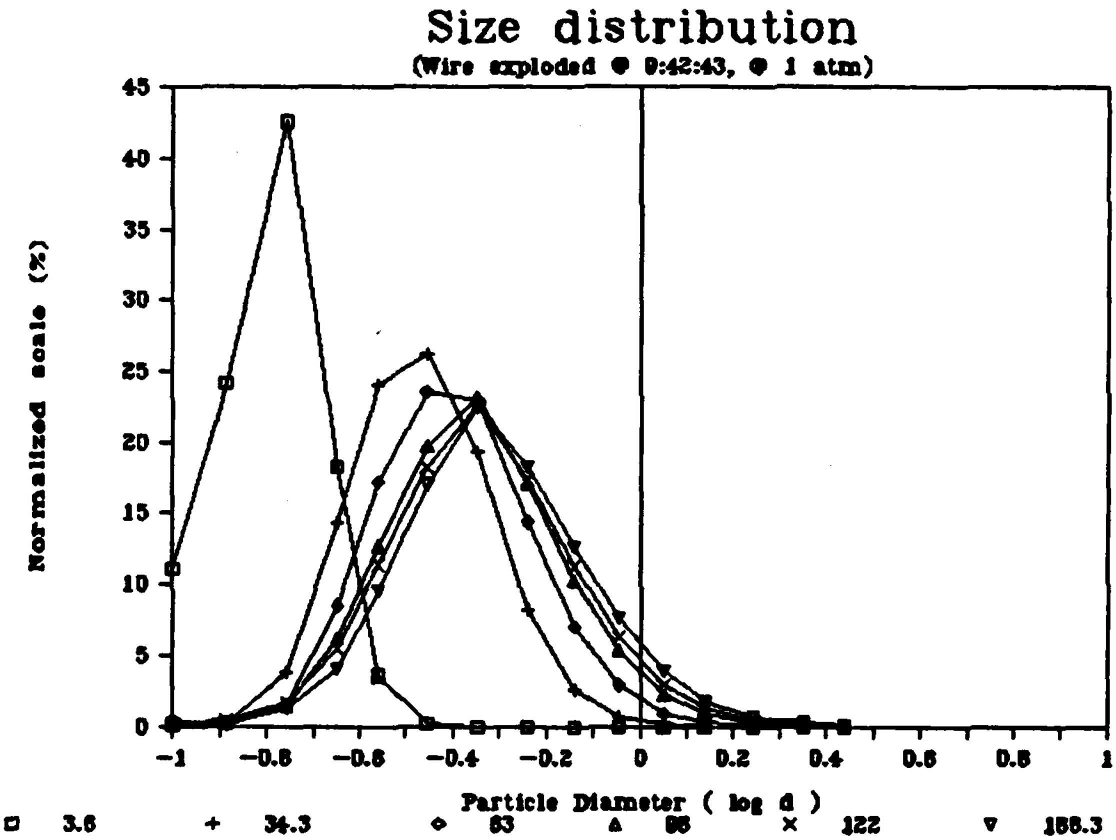

Figure 8. (Lower) Silver aerosol coagulated at 1.0 atmosphere for different times (values in seconds).

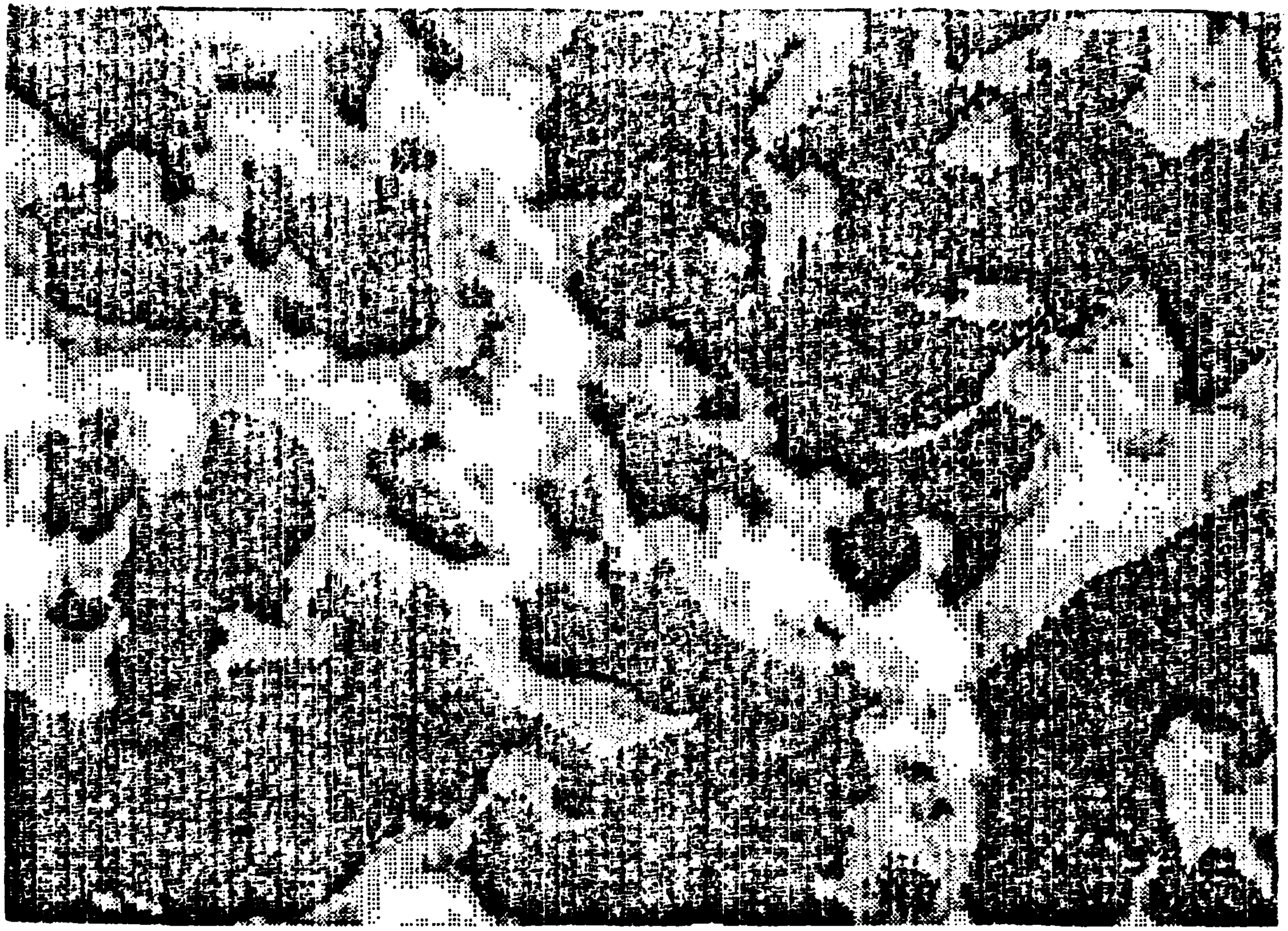

Figure 9. Digitized image of photomicrograph, printed on dot-matrix printer.

the aerosol, such as gold. This would have proved expensive for the exploratory work, although it remains a possibility for the later efforts.

In conjunction with the desire to have a metallic aerosol, it was also necessary to have a low residual electric charge on the particles so that electrically dominant forces would not influence the coagualation. The charges on numerous metal aerosols were measured with a Faraday cage filter, and a confirmation of the correlation between degree of charge and melting point was found. The higher melting point materials appear to become thermally ionized during the explosion, while the low melting point metals are dispersed without ionization.

In a fortuitous combination of low melting point and oxidation resistance, we have settled on the use of silver aerosols for the majority of the program but still have the options of both lower melting point and greater oxidation resistance available.

The first electron microscope (SEM) pictures of the particles show that a relatively uniform spherical primary particle is formed in the explosion (see Figure 10). The size and number density of these particles should be controllable with the explosion parameters, and we will attempt to describe their formation in terms of the condensation of material under the reduced pressure conditions.

It is also clear that the primary particles coagulate with time into dendritic shapes so that it becomes very difilcult to assign a characteristic diameter to the resulting particles. This has led to our consideration of the role of fractals in the description of the resulting growth.

We have also completed a series of measurements on the same wire, exploded under reduced pressure conditions. This series has been used as much to establish the sampling procedures as to obtain the particle information, but distinct differences are already apparent in the distributions. The formation of the dendritic structure is qualitatively different at 1.0 atmosphere from 0.01 atmosphere, but the difference is hard to put into words. Basically, the particles grown at 0.01 atmosphere appear smoother than at the higher pressures. It is our hope that the fractal analysis will pick this difference out mathematically and allow us to describe the particles with more precision.

Figure 10 shows the particles formed at 1 atmosphere after 20 seconds and 192 seconds of growth. The similarity of structure of the particles is clear, along with the difference in length of the largest dimension.

CONCLUSION

The first year's work on this project has proceeded at a slightly slower pace than anticipated in the proposal, but the progress has been accelerating as the period progressed. We had not anticipated the inclusion of the fractal analysis when the proposal was written, but it appears to be a most timely development for our understanding of the growth process.

The theoretical approaches to the problem are being refined as the data from the experiments becomes available, and fractal descriptions of types of growth processes are under development.

The experimental apparatus is about 90 percent complete, and most changes in it will be for operating convenience and consistency. The sampling techniques appear to be working as intended.

The results so far do show differences in the growth of aersols under normal and reduced pressures. The question of the quantitation of these differences is one of the first tasks awaiting us under the continuation of the effort.

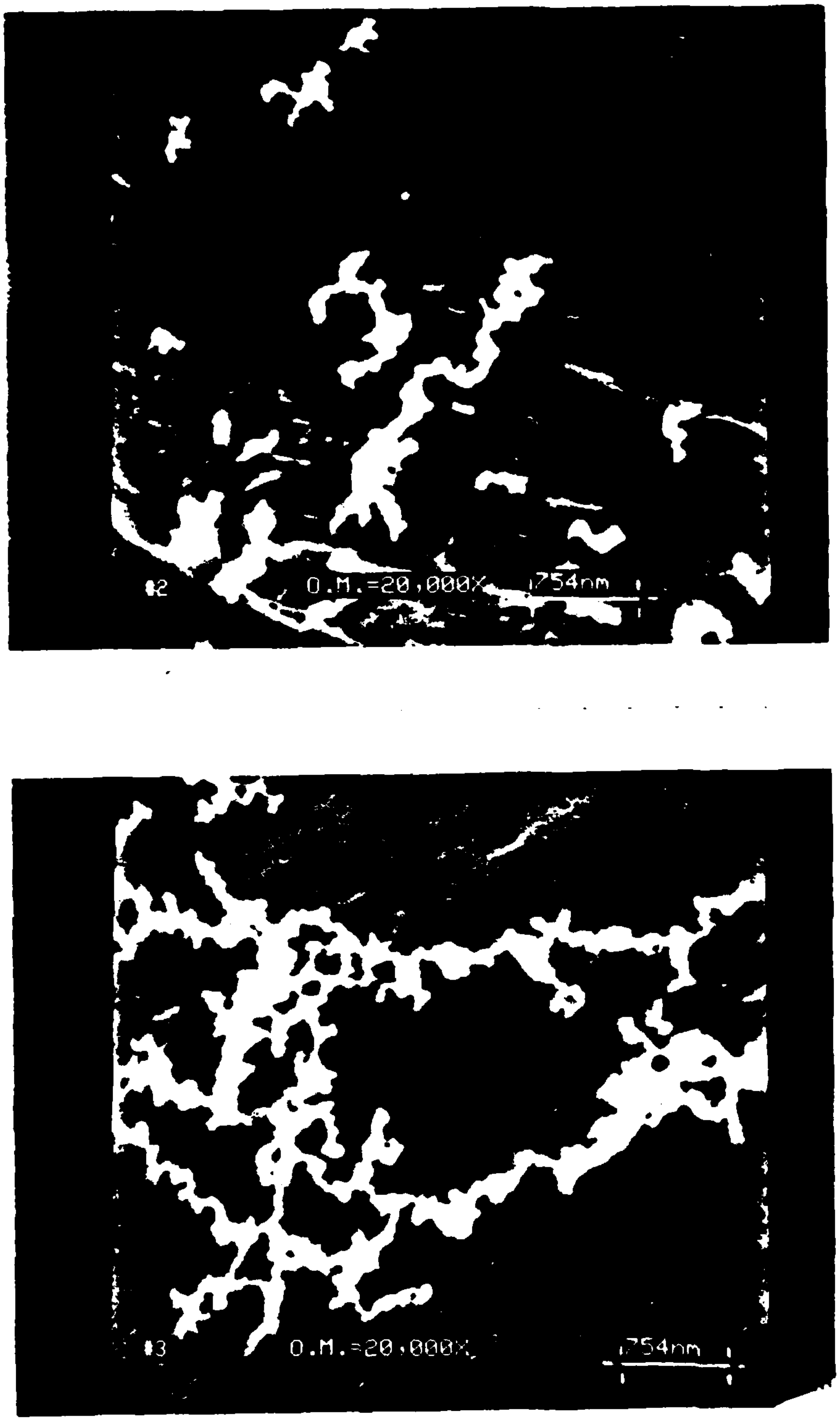

Figure 10. Electron micrographs of silver aerosol at 1.0 atmosphere. (Upper, 20 seconds of growth; lower, 192 seconds of growth.)

PUBLICATIONS

In preparation:

Pseudo-Self-Preserving Particle Size Distribution for Brownian Coagulation in the Transition Regime. A. S. Damle and P. A. Lawless. To be submitted to the JOURNAL OF AEROSOL SCIENCE.

Fractal Characterization of Particles Grown by Coagulation under Reduced Pressures. P. A. Lawless, P. C. Reist, and M. T. Hsieh. To be presented at CRDC Conference on Obscuration and Aerosol Research (June 1985).

PROFESSIONAL PERSONNEL

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